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*M.B. Chadwick, T. Kawano, M. White, R.
Nelson, Devlin, Fotiadis, P. Garrett, J.A. Becker*

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New yttrium evaluated cross sections and impact on $^{88}\text{Y}(n,2n)^{87}\text{Y}$ radchem

M.B. Chadwick¹, T. Kawano¹, M. White¹, R. Nelson¹, Devlin¹, Fotiadis¹
P. Garrett², J.A. Becker²

¹University of California, Los Alamos National Laboratory,
Los Alamos, New Mexico 87545, USA

²University of California, Lawrence Livermore National Laboratory,
Livermore, California 94550, USA

Abstract

We evaluate new $n+^{89}\text{Y}$ radchem cross sections using recent LANSCE/GEANIE measurements and GNASH nuclear model calculations, together with previous measurements at Livermore by Dietrich et al. A quantification of margins and uncertainties (QMU) analysis leads to evaluated cross sections for the $(n,2n)$ population of the ^{88}Y ground state and $m1$, $m2$ isomers, together with uncertainties. Our new results agree with historic radchem database cross sections within a few percent below 15 MeV (with larger differences above 15 MeV), and they therefore provide a validation of the historic Arthur work that is used in LANL simulation codes.

Since the $(n,2n)$ cross sections to the ^{88}Y g.s. and $m1$, $m2$ isomers impact the average $^{88}\text{Y}(n,2n)^{87}\text{Y}$ cross section at leading-order, we determine the new 14.1 MeV average $^{88}\text{Y}(n,2n)^{87}\text{Y}$ cross section (crucially important for radchem). Our new 14 MeV average $^{88}\text{Y}(n,2n)^{87}\text{Y}$ cross section is 1107 mb ($\pm 4\%$) which agrees with the value obtained from the historic Arthur cross section data to 0.7 %.

1 Introduction

Yttrium plays a central role in radiochemical diagnostics of fusion yield. For this, accurate cross sections are needed, and in the 1970s Ed Arthur developed (Arthur, 1977) a set of yttrium cross sections – the pathways evaluated in this ‘historic’ database are shown in Figure 1. Use of both experimental data, where available, and model calculations, was made. A key quantity for interpreting radchem data is the 87/88 yttrium

ratio, which in leading order for low fluences depends upon the average $^{88}\text{Y}(n,2n)^{87}\text{Y}$ cross section. In leading order (Jungman, 2002) the average cross section is given by

$$\sigma_{88 \rightarrow 87}(n, 2n) = br_1 \sigma_{88 \rightarrow 87}^{g.s.}(n, 2n) + br_2 \sigma_{88 \rightarrow 87}^{m1}(n, 2n) + br_3 \sigma_{88 \rightarrow 87}^{m2}(n, 2n) \quad (1)$$

where br_1 , br_2 , br_3 , are the branching ratio fractions for $(n, 2n)$ reactions on ^{89}Y going to the ground, $m1$, and $m2$ states. In this low-fluence limit, the measured (second-order) $N(87\text{Y})/N(88\text{Y})$ ratio of atoms of yttrium isotopes is given by

$$N(87\text{Y})/N(88\text{Y}) = (1/2)\sigma_{88 \rightarrow 87}(n, 2n)\Phi \quad (2)$$

Where Φ is the neutron fluence. The factor of $1/2$ can be understood intuitively as follows. At time zero, the initial amount of ^{88}Y is zero; at the end of the irradiation it is $N(88\text{Y})$; and on average during the irradiation the amount of ^{88}Y that can be converted to ^{87}Y via $(n,2n)$ reactions is $1/2N(88\text{Y})$.

In the present work, the GEANIE measurement from a LLNL-LANL collaboration (Garrett *et al.*, 2003) provides a comprehensive description of the population of the ^{88}Y ground state and $m1$ isomer: all significant gamma-ray decays that populate these states are measured. Thus, the principal role played by GNASH nuclear theory is to predict the unmeasured contributions due to neutron sidefeeding (i.e. neutron emission that leads to the population of the ground state and isomeric state directly, without gamma-ray decay), to augment the GEANIE measurements. In order to build confidence in the accuracy of the calculations, we also compare the calculated gamma-ray cross sections for feeding the ground-state and isomers with the GEANIE measurements.

In the case of the $m2$ isomer, GEANIE does not measure a significant fraction of the decays that feed it, and therefore cannot be used here however, fortunately the $m2$ isomer production (and the $m1$, but not the ground state) were independently measured by Dietrich *et al* at 14.1 MeV, and the present work (like Arthurs historic work) makes use of the Dietrich data.

Many previous measurements exist for the total $^{89}\text{Y}(n,2n)^{88}\text{Y}$ cross section. These provide an important constraint on the sum of the evaluated $(n,2n)$ cross sections to the ground state, and $m1$, $m2$ isomers. These data are shown later in this report.

2 Nuclear theory and modeling methods

The GNASH nuclear reaction code (Young *et al.*, 1998) was used to model the neutron reactions on ^{89}Y . An earlier version of this code was used by Arthur in his original yttrium cross section work in the 1970s and early 1980s. New advancements in the calculational methods compared to the early work include a more modern treatment of preequilibrium reaction processes; a modeling of gamma-ray decay strength functions using the Kopecky-Uhl formalism; and use of more modern nuclear spectroscopy information describing low-lying levels and their gamma-ray decay branching fractions.

However, we have maintained some of the original modeling details that Arthur developed. The spherical optical model we used was based on Arthur's original work, since it was carefully developed to model known elastic and total neutron scattering data, and S-wave strength functions. The optical model for charged-particle emission, particularly proton emission, was validated against measured p+Sr reaction cross section and (p,n) data.

Importantly, we have followed Arthur's original assumption, based on an earlier work (Dietrich *et al.*, 1974) at Livermore, for the ^{88}Y spectroscopy. Specifically, we have assumed there exists a 7+ state that is part of a particle-hole band of states, 8+, 7+, 6+, 5+, formed from the coupling of $(g_{9/2})$ proton-particle, $(g_{9/2})$ neutron-hole states, of which the 8+ lowest-lying state is the m2 isomer of interest. Even though the 7+ state has not been measured, it is highly probable that it exists. This then allows the states 6+, 5+ *etc* to gamma-ray decay at a significant level through the 7+ state into the 8+ m2 isomeric state. With this assumption, GNASH calculations are in good agreement with the measurements.

3 Evaluated Cross Sections

3.1 $^{89}\text{Y}(\text{n},2\text{n})$ to the ground-state

As a validation test, we have compared the GEANIE measurement of the sum of gamma-rays feeding the ground-state, compared with the calculated sum. Agreement was seen to be fairly good (at the 20 percent level), providing some confidence in the calculational methods (Chadwick, 2003). In our best estimate of the total cross section to the ground state, the contribution from gamma-ray feeding is taken from the GEANIE data, not the GNASH calculation.

The GNASH calculation is used to determine the unmeasured neutron sidefeeding component (with an estimated calculational uncertainty of 20%). When this is done, and added to the measured GEANIE gamma-ray feeding, the result is shown in Fig. 1 (filled circles). These GEANIE-GNASH best results are also compared with the historic Arthur evaluation (red dashed line). No other measurements previously existed for this ground state production cross section.

Our new evaluation of this cross section is shown as a solid line - based upon information from GEANIE/GNASH, from Dietrich measurements, and from the constraint of the total (n,2n) cross section which is known rather precisely. Further details on this evaluation are given later. The blue triangle at 14.1 MeV is our evaluated best estimate cross section - see the appendix for how this was determined.

3.2 $^{89}\text{Y}(\text{n},2\text{n})$ to the m1 isomer

For validation purposes we compared the GEANIE measurement of the sum of 4 gamma-rays feeding the m1 isomer, compared with the calculated sum. Agreement was again seen to be fairly good, at the 20 percent level (Chadwick, 2003). In our best

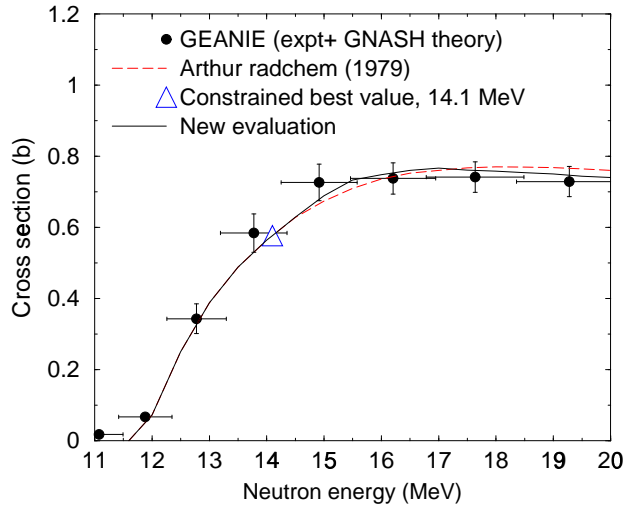


Figure 1: The $^{89}\text{Y}(n,2n)$ cross section to the ground state.

estimate of the total cross section to the m1 isomer, the contribution from gamma-ray feeding is taken from the GEANIE data, not the GNASH calculation.

The GNASH calculation is used to determine the unmeasured neutron sidefeeding component. When this is done (with a 20% uncertainty assigned), and added to the measured GEANIE gamma-ray feeding, the result is shown in Fig. 2 (filled circles). These GEANIE-GNASH results are also compared with the historic Arthur evaluation (red dashed line).

We also compare the GEANIE-GNASH results with other measurements for the m1 cross section. At 14.1 MeV, our results are consistent with the Dietrich *et al.* measurement. There are also other measurements by Eapen and by Monnard that are inconsistent with the present work and with Dietrich's data.

Our new evaluation of this cross section is shown as a solid line - based upon information from GEANIE/GNASH, from Dietrich measurements, and from the constraint of the total (n,2n) cross section which is known rather precisely. Further details on this evaluation are given later. The blue triangle at 14.1 MeV is our evaluated best estimate cross section - see the appendix for how this was determined.

3.3 $^{89}\text{Y}(n,2n)$ to the m2 isomer

Just one weak gamma-ray feeding the m2 isomer was measured by GEANIE, thus rendering the GEANIE experiment inadequate for inferring information on this m2 cross section. We compared the GEANIE measurement of this one gamma-ray feeding the m2 isomer, with the calculated cross section. Agreement was again seen to be fairly good, at the 20 percent level (Chadwick, 2003).

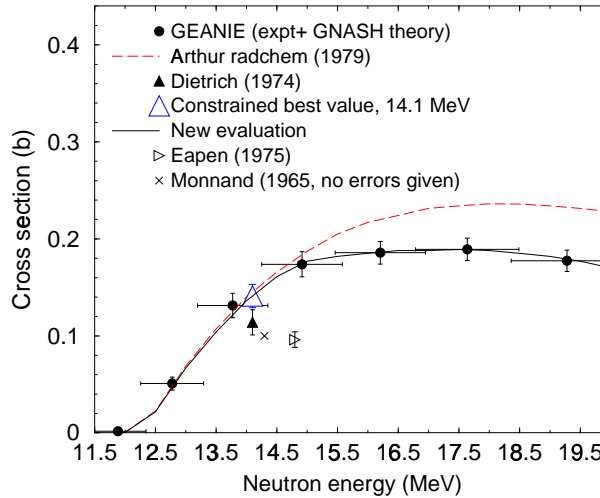


Figure 2: The $^{89}\text{Y}(n,2n)$ cross section to the m1 isomer.

Fortunately the m2 cross section was measured by Dietrich, see Fig. 3. We show this measurement compared with the historic Arthur evaluation (red dashed line). Agreement is seen to be good. Other measurements by Eapen and by Monnard are here consistent with Dietrich's data, but measurements by Van Zeist and by Garg are seen to be inconsistent.

Our new evaluation of this cross section is shown as a solid line - based upon information from GEANIE/GNASH (for the other g.s. and m1 channels), from Dietrich measurements, and from the constraint of the total $(n,2n)$ cross section which is known rather precisely. Further details on this evaluation are given later. The blue triangle at 14.1 MeV is our evaluated best estimate cross section - see the appendix for how this was determined.

4 Evaluation Procedure

To evaluate new $^{89}\text{Y}(n,2n)$ cross sections to the ground state and m1, m2 isomers, we make use of the GEANIE-GNASH results for the ground and m1 states; the Dietrich 14.1 MeV data for the m1 and m2 states; and the constraint that the total $(n,2n)$ cross section is known accurately at 14.1 MeV. We start by evaluating the best-estimate cross sections at 14.1 MeV, and then proceed to all incident energies.

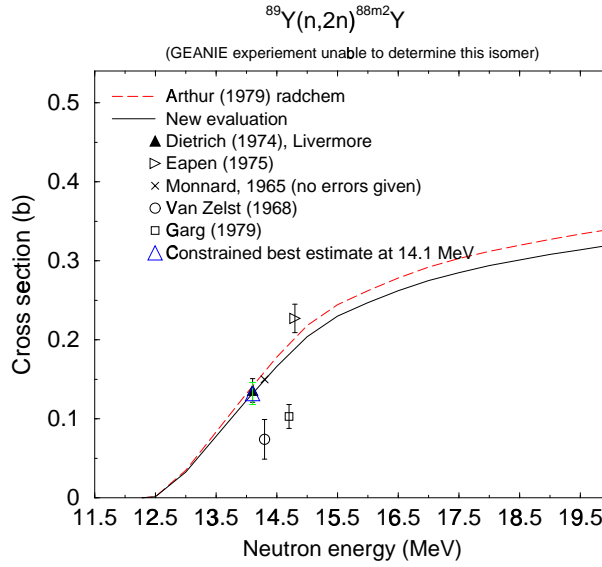


Figure 3: The $^{89}\text{Y}(n,2n)$ cross section to the m2 isomer.

4.1 Cross section at 14.1 MeV

We have included the Livermore Dietrich et al. 14.1 MeV data in our evaluation because we believe them to be reliable. We do not have such confidence in the other measurements by Eapen, Monnard, Van Zeist, and Garg, and have therefore not used them (though we have shown them in the earlier figures).

Based on a wealth of data for the total $^{89}\text{Y}(n,2n)^{88}\text{Y}$ cross section, Don Barr evaluates this cross section as $845 \text{ mb} \pm 2\%$ at 14.1 MeV (Barr, 2002). Note that this is based on the LANL measurement methodology (counting methods consistent with those used after a test). The $(n,2n)$ data are shown in Fig. 4. This figure also shows Arthur's historic evaluation (dashed red line), and our new evaluation (bold solid line) which is described in more detail below.

We have undertaken a Bayesian analysis of these data at 14.1 MeV, including the above constraint from the total $(n,2n)$ cross section, giving the following results (see Appendix): These best-estimate evaluated results at 14.1 MeV are shown in figs.1,2,3 for the g.s., m1, and m2 isomers, as blue triangles. Clearly they are in good agreement with the historical values (red dashed curves) of Arthur. At 14.1 MeV the deviations compared to Arthur for the ground state, and m1, m2 isomers, are 0.02, -2.83, -6.52 % respectively. Therefore, our work has provided a validation of the historical evaluation by Arthur. The total $(n,2n)$ cross section we obtain at 14.1 MeV is 850 mb - slightly higher than Barr's evaluation of 845 mb ($\pm 2\%$) because of the higher GEANIE data for the ground state (see the Appendix), but well within the 2% uncertainty.

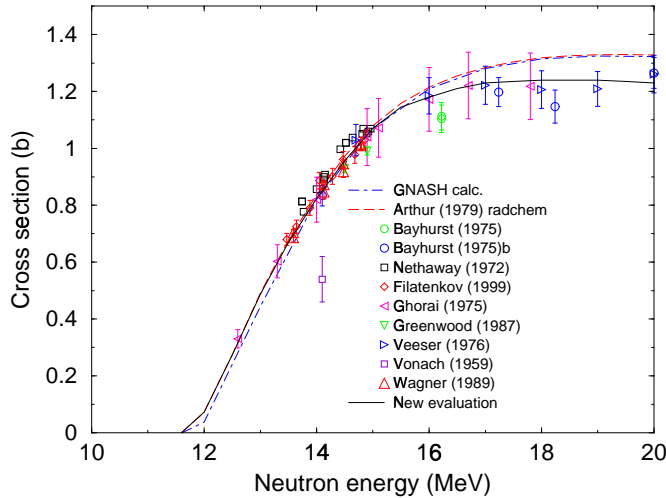


Figure 4: The $^{89}\text{Y}(n,2n)$ total cross section (sum of ground, m1 and m2).

4.2 Cross sections from threshold to 20 Mev

Arthur's original evaluation was based on a calculation similar to the present GNASH calculation. Physically, the evaluated results are expected to be strongly correlated at nother eutron energies because values at all energies are based on a common set of nuclear level densities, transmission coefficients, *etc.* Thus, we have obtained re-evaluated cross sections for the ground, and m1, m2 isomers by scaling the historic Arthur cross sections by 0.02, -2.83, -6.52 % below 15 MeV. At higher energies (15-20 MeV) we have based our new evaluations on the new GEANIE/GNASH data, together with the total (n,2n) constrain. The new evaluations are shown as solid black lines in Figs. 1,2,3,4.

5 Impact on average $^{88}\text{Y}(n,2n)^{87}\text{Y}$ cross section at 14.1 MeV.

As indicated in the Introduction, for low fluences the average cross section for $^{88}\text{Y}(n,2n)^{87}\text{Y}$ is given by Eq. 1. This allows us to compare the average cross section based on the new evaluation with that based on the historic Arthur evaluation at 14.1 MeV.

The (n,2n) cross section from the ^{88}Y ground state was measured in a heroic measurement by Prestwood, Nethaway *et al.*, in a LLNL-LANL collaboration (Prestwood, 19??). The (n,2n) cross section from the m1 and m2 isomers, as targets, have not been measured, and were determined by Arthur using GNASH model calculations. We have adopted (unchanged) Arthur's original evaluation for these ^{88}Y target cross

sections.

We need to estimate uncertainties on the $^{88}\text{Y}(n, 2n)^{87}\text{Y}$ cross sections, which were obtained from the historic Arthur evaluation. For the ground state target, we estimate 3.6% uncertainty since this is the quoted experimental uncertainty (Barr, 2002) on the measured data (1129 mb pm 40 mb). Even though the n,2n cross sections from ^{88}Y m1 and m2 targets remain unmeasured, we expect that the historic Arthur model predictions are reliable. The reason is that the GNASH model calculations were first performed for the ^{88}Y ground state, where measurements do exist. Since calculation and experiment agree well here, the same model input parameters are then used for the isomer-target calculations. The excitation energy effect of the isomer target is analogous to an increased incident energy, so that predicting the cross sections for isomer targets is similar to predicting excitation functions for energies above 14.1 MeV, after calculation and measurement agree at 14.1 MeV – something that can be done fairly reliably. The m1 isomer is of low spin (1+), with an excitation energy (0.393 MeV), and thus we think that the GNASH model calculation extrapolations from the known ground state target to the unmeasured m1 isomer are well constrained, and we estimate a m1 target $^{88}\text{Y}(n, 2n)^{87}\text{Y}$ uncertainty of 5%. However, the m2 isomer is of high spin (8+), with an excitation energy (0.675 MeV), and the GNASH model calculation extrapolations from the known ground state target to the unmeasured m2 isomer are less well constrained. Interestingly, instead of being higher than the g.s, or m1 target (as one would expect from just energy considerations) the predicted m2 target is actually lower, because of spin-dependent level density phase space effects in the n,2n reaction. We have explored different modeling assumptions to estimate an uncertainty on this prediction, resulting in an estimated m2 target $^{88}\text{Y}(n, 2n)^{87}\text{Y}$ uncertainty of 15%. All these uncertainties are 1-sigma.

In Equation 1, the best-estimate branching ratios to the gs, m1 and m2 are 0.679, 0.165, and 0.155 respectively (compared to historic Arthur values of 0.669, 0.168, and 0.163). The 14.1 MeV $^{88}\text{Y}(n, 2n)$ cross sections from the ground, m1, and m2 states as targets are 1123 mb \pm 40 mb, 1238 mb \pm 62 mb, 856 mb \pm 128 mb, respectively, as taken from the Arthur ^{88}Y target evaluation unchanged.

The Appendix details the calculated average (n,2n) cross section. Our results for the average $^{88}\text{Y}(n, 2n)^{87}\text{Y}$ cross sections, using Eq. 1, are as follows:

Historic = 1099 mb (no uncertainties given)

New evaluation = 1107 \pm 4% mb

Thus, the average $^{88}\text{Y}(n, 2n)^{87}\text{Y}$ cross section based on our new evaluation is extremely close to that based on the historic data, the difference being 0.7%.

6 Conclusions

Our new yttrium evaluated cross sections, based on recent LANSCE/GEANIE gamma-ray measurements augmented with nuclear model calculations for unmeasured contributions, have been shown to be close to Arthur's historic evaluated cross sections. The new results are incorporated into an ENDF formatted file that has been made

available in different formats for Laboratory simulation codes. This paper has also provided uncertainties in the cross sections. UCRL-TR-202884

We have explored the impact of these new cross sections on the second order $^{87/88}\text{Yttrium}$ ratio for fluence determinations. Compared to the historic data, the new results are 0.7% different for a monoenergetic 14.1 MeV fluence. Similar comparisons for other (more realistic) neutron spectra, other than the 14.1 MeV value given above, are of great practical interest but must be presented elsewhere.

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Appendix: Calculation of Uncertainty in the Averaged Cross Section

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In the mono-energetic neutron environment, the averaged $^{88}\text{Y}(n, 2n)$ cross section $\sigma_{88 \rightarrow 87}(n, 2n) = \bar{\sigma}$ is given by Eq. (1). We rewrite this as

$$\bar{\sigma} = \frac{\alpha_0}{\sigma_{tot}}\beta_0 + \frac{\alpha_1}{\sigma_{tot}}\beta_1 + \frac{\alpha_2}{\sigma_{tot}}\beta_2, \quad (3)$$

where α_i is the $(n, 2n)$ cross section of ^{89}Y , the index i stands for the produced state ($i = 0$ for the ground state of ^{88}Y , $i = 1$ for the first, and $i = 2$ for the second meta-stable state, respectively), β_i 's are the $^{88g}\text{Y}(n, 2n)$, $^{88m_1}\text{Y}(n, 2n)$, and $^{88m_2}\text{Y}(n, 2n)$ reaction cross sections, σ_{tot} is the total $^{89}\text{Y}(n, 2n)$ cross section which is equal to $\sum \alpha_i$.

The α_i values in Eq. (3) can be estimated by considering nuclear model calculations and experimental data available. Those information is expressed by a pair of value and its error — $\alpha_i \pm e_i$. The sum of those cross sections $\sigma_{tot} = \sum \alpha_i$ has a very small uncertainty $e \rightarrow 0$ where e is the uncertainty of σ_{tot} , since this value is experimentally unambiguous, as described in the text. Our estimated value is $\sigma_{tot} = 845 \text{ mb} \pm 2\%$.

The Bayesian method tells us the best estimates of α_0 , α_1 , and α_2 when the condition $\sum \alpha_i = 845 \text{ mb}$ is provided. This gives the same equation as a least-squares solution with constraint. The prior parameter vector $\mathbf{s}^{(1)}$ contains the cross sections $\mathbf{s}^{(1)} = (\alpha_0, \alpha_1, \alpha_2)^t$, where t stands for transpose. According to the Bayesian method, the posterior parameter vector $\mathbf{s}^{(2)}$ is given by

$$\mathbf{s}^{(2)} = \mathbf{s}^{(1)} + VD^t (DVD^t + e)^{-1} (\sigma_{tot} - D\mathbf{s}^{(1)}), \quad (4)$$

$$P = V - VD^t (DVD^t + e)^{-1} DV, \quad (5)$$

where V is the covariance of $\mathbf{s}^{(1)}$, and P is the covariance of $\mathbf{s}^{(2)}$. The matrix D is called a design matrix which connects parameters and observables. Because our function which relates the parameter α_i with σ_{tot} is $\sigma_{tot} = \sum \alpha_i$, the D matrix becomes a simple vector,

$$D\mathbf{s} = (1, 1, 1) \begin{pmatrix} \alpha_0 \\ \alpha_1 \\ \alpha_2 \end{pmatrix} = \sum \alpha_i. \quad (6)$$

The covariance V contains uncertainties in the α 's as

$$V = \begin{pmatrix} e_0^2 & & \\ & e_1^2 & \\ & & e_2^2 \end{pmatrix} \quad (7)$$

We estimated the prior vector $\mathbf{s}^{(1)}$ by considering the experimental data and the model calculation with GNASH, which were

$$\mathbf{s}^{(1)} = \begin{pmatrix} 625 \\ 144 \\ 136 \end{pmatrix} \pm \begin{pmatrix} 53 \\ 13 \\ 15 \end{pmatrix} \quad \text{mb.} \quad (8)$$

From Eqs. (4) and (5), we obtain

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i	α [mb]	Error [mb]	Correlation [%]		
0	577	18	100		
1	141	12	-62	100	
2	132	14	-74	-6	100

The sum of α 's in the table above is slightly larger than the experimental σ_{tot} value because of e in Eqs. (4) and (5).

The uncertainty in $\bar{\sigma}$ of Eq. (3) is given by the error propagation from the uncertainties in α_i and β_i , which is given by

$$\delta\bar{\sigma} = CXC^t, \quad (9)$$

where C is the sensitivity matrix, and X is the covariance of parameters. The sensitivity matrix is calculated as

$$\begin{aligned} C &= \left(\frac{\partial\bar{\sigma}}{\partial\alpha_0}, \frac{\partial\bar{\sigma}}{\partial\alpha_1}, \frac{\partial\bar{\sigma}}{\partial\alpha_2}, \frac{\partial\bar{\sigma}}{\partial\beta_0}, \frac{\partial\bar{\sigma}}{\partial\beta_1}, \frac{\partial\bar{\sigma}}{\partial\beta_2} \right) \\ &= \frac{1}{\sigma_{tot}} (\beta_0, \beta_1, \beta_2, \alpha_0, \alpha_1, \alpha_2). \end{aligned} \quad (10)$$

We can assume that there are no correlations between α 's and β 's, therefore the covariance matrix X can be decomposed into two sub-matrices,

$$X = \begin{pmatrix} P & \\ & Q \end{pmatrix}, \quad (11)$$

where P is the covariance of α 's given by Eq. (5), and Q is the covariance of β values. Note that we do not need to have a sensitivity of σ_{tot} in Eq. (10), because this is implicitly taken into account through the covariance P of α 's.

The β_i values which are $^{88g}\text{Y}(n, 2n)$, $^{88m_1}\text{Y}(n, 2n)$, and $^{88m_2}\text{Y}(n, 2n)$ reaction cross sections at 14.1 MeV were estimated in the main text, and we assumed the following covariance matrix Q .

i	β [mb]	Error [mb]	Correlation [%]		
0	1123	40	100		
1	1238	62	100	100	
2	856	128	10	10	100

With those β_i values the calculated $\bar{\sigma}$ given by Eq. (3) and its uncertainty by Eq. (9) are $\bar{\sigma} = 1107 \text{ mb} \pm 45 \text{ mb}$.

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